

# A Concurrent Multiscale Micromorphic Molecular Dynamics. Part I. Theoretical Formulation

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(Dated: March 10, 2014)

Based on a novel concept of multiplicative multiscale decomposition, we have derived a multiscale micromorphic molecular dynamics (MMMD) to extend the (Andersen)-Parrinello-Rahman molecular dynamics to mesoscale and macroscale. The multiscale micromorphic molecular dynamics is a con-current three-scale particle dynamics that couples a fine scale molecular dynamics, a mesoscale particle dynamics of micromorphic medium, and a coarse scale nonlocal particle dynamics of nonlinear continuum together. By choosing proper statistical closure conditions, we have shown that the original Andersen-Parrinello-Rahman molecular dynamics can be rigorously formulated and justified from first principle, and it is a special case of the proposed multiscale micromorphic molecular dynamics. The discovered multiscale structure and the corresponding multiscale dynamics reveal a seamless transition from atomistic scale to continuum scale and the intrinsic coupling relation among them. The proposed MMMD can be used to solve finite size nanoscale science and engineering problems with arbitrary boundary conditions.

PACS numbers: 02.70.-c, 07.05.Tp, 31.15.At, 31.15.xv, 31.15.ee

## I. INTRODUCTION

Thirty some years ago, in his seminal work Andersen [1] first proposed an isoenthalpic-isobaric ensemble of molecular dynamics (MD) allowing the volume a cubic lattice cell to vary. Subsequently, Parrinello and Rahman [2, 3] elegantly extended Andersen's formalism to anisotropic cases allowing both the volume and the shape of a molecular dynamics (MD) cell to vary. Since then the (Andersen)-Parrinello-Rahman (APR) molecular dynamics has become the standard molecular dynamics method in simulations of structural transformation and phase transitions.

However, the APR-MD approach has not been thoroughly understood, and this is reflected in both its physical foundation as well as how to extend it beyond the restriction of periodic boundary condition so that it can bridge up to macroscale or continuum scale. Moreover, the APR-MD lagrangian has been viewed as an ad hoc choice, as Parrinello and Rahman commented in [3], “..... Whether such a Lagrangian is derivable from first principles is a question for further study; its validity can be judged, as of now, by the equations of motion and the statistical ensembles that it generates. ....”

In recent years, there have been renewed interests in revising APR molecular dynamics, e.g. [4–7], which attempted to extend APR MD to non-equilibrium condition or to macroscale simulation. On the other hand, there have been many efforts to formulate multiscale coarse-grained molecular dynamics, among them [8–13] are probably most rigorous approaches.

One of the objectives of the present work is to revise or generalize APR molecular dynamics so that it can lead to a rigorous macroscale nonlinear continuum mechanics, e.g. [7, 14]. By doing so, one hopes to achieve a multiscale molecular dynamics that is not necessary to impose the periodic boundary condition.

In this paper, we revisit the topic again in an attempt to accomplish the following two objectives:

- (1) Derive the original (Andersen)-Parrinello-Rahman (APR) Lagrangian from first principle, and
- (2) Extend the single-cell APR molecular dynamics to a multiscale continuum-molecular dynamics so that it is capable of simulating lattice dynamic motions in multiple cells without imposing periodic boundary condition, and bridging molecular dynamics and nonlinear continuum mechanics from first principle.

The paper is arranged into seven Sections. In the second Section, we discuss the multiscale decomposition that includes both micromorphic decomposition (multiplicative decomposition) and the additive decomposition. In the third section, we discuss the random variables and their statistical closure. Section 4 restates the original PR-MD formulation. In Section 5, we derive the general formulae for the multiscale micromorphic molecular dynamics; and in Section 6, we discuss a coarse-grain approach to approximate the multiscale molecular dynamics when the fine scale

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dynamics are turned off — that is how to obtain a rigorous coarse grained molecular dynamics. Finally, in Section 7, we close the presentation by making few comments and remarks.

## II. MULTISCALE DECOMPOSITIONS

The key to extend the APR molecular dynamics to continuum scale is how to decompose the material displacement field into different scales, and so that we can establish the coupled dynamics equations of displacement field at different scales. In fact, the main reason why the PR-MD has been successful is its ingenious multiscale decomposition of the displacement field that reveals connections between mesoscale thermodynamics with atomistic motion at the fine scale.

Before we construct any multiscale PR-MD, it would be instrumental to discuss the multiscale decomposition of the material displacement field first. In the literature, there are two types of multiscale decompositions in molecular dynamics simulations: (1) Micromorphic decomposition, or multiplicative decomposition, and (2) Wagner-Liu's additive decomposition.

To start with, we first assume that the crystalline material has periodic lattice structure and its deformation is locally uniform so that we can discretize the entire domain into many cells,  $\alpha = 1, 2, \dots, N_c$ , such that at macroscale the coarse continuum deformation in each cell is spatially uniform.

We assume that for the  $i$ -th atom inside the  $\alpha$ -th cell, its absolute displacement follows the following micromorphic decomposition,

$$\mathbf{r}_i(t) = \mathbf{r}_\alpha(\mathbf{R}, t) + \mathbf{r}_{\alpha i}(\mathbf{R}_i, t), \quad i = 1, 2, \dots, N_a$$

where  $\mathbf{r}_\alpha$  is the spatial position of the center of mass in the  $\alpha$ -th cell, which may be identified as or a function of coarse scale coordinate  $\mathbf{R}$ . When  $t = 0$ , we have

$$\mathbf{R}_i = \mathbf{R}_\alpha + \mathbf{R}_{\alpha i}, \quad i = 1, 2, \dots, N_a \quad (1)$$

Suppose there are  $N_\alpha$  atoms in the  $\alpha$ -th cell, the center of mass in the  $\alpha$ -th cell at the referential configuration of the medium  $\Omega_\alpha(0)$  is

$$\mathbf{R}_\alpha = \frac{1}{\sum_i m_i} \sum_i m_i \mathbf{R}_i, \quad \forall \mathbf{R}_{\alpha i} \in \Omega_\alpha(0) \quad (2)$$

We can then justify the decomposition Eq. (1), Note that Eq. (2) is not an additional constraint, but an average property. That is the weighted average micromorphic coordinates of a ensemble system is a single coordinate field, i.e. coarse scale coordinate. Whereas the average of the fine scale coordinates is zero,

$$\sum_i m_i \mathbf{R}_{\alpha i} = 0.$$

Similar decomposition is hold in the current configuration  $\Omega(t) = \bigcup_\alpha \Omega_\alpha(t)$ , i.e.

$$\mathbf{r}_i = \mathbf{r}_\alpha + \mathbf{r}_{\alpha i}, \quad \text{and} \quad \mathbf{r}_\alpha = \frac{1}{\sum_i m_i} \sum_i m_i \mathbf{r}_i \quad (3)$$

By definition,

$$\sum_i m_i \mathbf{r}_{\alpha i} = 0, \quad \text{and} \quad \sum_i m_i \dot{\mathbf{r}}_{\alpha i} = 0. \quad (4)$$

Note that  $\mathbf{r}_\alpha$  is the position of center of mass of the  $\alpha$ -cell, and it can be used to represent the displacement field in the coarse scale dynamics equations, which is one of the focus of this work.

We consider the so-called Micromorphic Decomposition. In the micromorphic theory, a micro deformation tensor is assigned to each cell, i.e.  $\boldsymbol{\phi}^\alpha = \phi_{ij}^\alpha \mathbf{e}_i \otimes \mathbf{e}_j$ , which characterizes the microstructure of the cell. In the PR-MD theory, the relative position of each atom in a deformed cell is related to the micro-deformation tensor by,

$$\mathbf{r}_{\alpha i}(\mathbf{R}_i, t) := \boldsymbol{\phi}_\alpha(t) \cdot \mathbf{S}_{\alpha i}(t),$$

where  $\mathbf{S}_{\alpha i}(t)$  is a statistical field variable that is a function of time. This is in contrast to the deterministic continuum mechanics, in which  $\mathbf{R}_{\alpha i}$  is a fixed vector in the referential configuration. A simple way to distinguish the deterministic referential coordinate  $\mathbf{R}_i$  and statistical coordinate  $\mathbf{S}_i$  is to set

$$\mathbf{S}_i(t) = \mathbf{R}_i + \mathbf{T}_i(t), \quad \text{and} \quad \langle \mathbf{T}_i(t) \rangle = 0, \quad \text{or} \quad \langle \mathbf{S}_i(t) \rangle = \mathbf{R}_i,$$

where the operator  $\langle \cdot \rangle$  is temporal average operator, which will be discussed in Section 3 in details.

The Second Multiscale Decomposition is the Wagner and Liu's additive decomposition, in which the following additive decomposition of the displacement field is introduced,

$$\mathbf{u}_i(t) = \bar{\mathbf{u}}_i(\mathbf{R}_i, t) + \mathbf{u}'_i(t) \quad (5)$$

where  $\mathbf{u}'_i$  is the fine scale displacement of the  $i$ -th atom, and  $\bar{\mathbf{u}}_i$  is the value of the coarse scale displacement of the atom  $i$  that may be determined by continuous displacement field, e.g. the finite element interpolation field,

$$\bar{\mathbf{u}}(\mathbf{R}_i) = \sum_I N_I(\mathbf{R}) \Big|_{\mathbf{R}=\mathbf{R}_i} \mathbf{d}_I$$

where  $N_I(\mathbf{R})$  is the finite element interpolation function. Following this construction, intuitively one may define and the fine scale relative displacement field in a micromorphic displacement field as

$$\mathbf{r}'_{\alpha i} = \phi_{\alpha}(t) \mathbf{S}_i - \mathbf{F}(\mathbf{R}_{\alpha}, t) \cdot \mathbf{R}_{\alpha i} \quad (6)$$

where  $\mathbf{F}(\mathbf{R}_{\alpha}, t)$  is the coarse scale deformation gradient, which can be calculated as,

$$\mathbf{F}(\mathbf{R}_{\alpha}, t) = \frac{\partial \mathbf{r}}{\partial \mathbf{R}} \Big|_{\mathbf{R}=\mathbf{R}_{\alpha}} = \sum_I \left( \mathbf{d}_I \otimes \frac{\partial N_I}{\partial \mathbf{R}} \right) \Big|_{\mathbf{R}=\mathbf{R}_{\alpha}} + \mathbf{I}^{(2)}$$

This definition (6) is consistent with (5), e.g.

$$\mathbf{r}_{\alpha i} = \bar{\mathbf{r}}_{\alpha i} + \mathbf{r}'_{\alpha i},$$

if only the Cauchy-Born rule can be applied for the coarse scale displacement, i.e.

$$\bar{\mathbf{r}}_{\alpha i} \approx \mathbf{F}(\mathbf{R}_{\alpha}, t) \cdot \mathbf{R}_{\alpha i}, \quad \text{and} \quad \mathbf{F}(\mathbf{R}_{\alpha}, 0) = \mathbf{I}^{(2)}.$$

In the rest of the paper, we assume that the Cauchy-Born rule approximation stands at coarse scale, unless it is stated otherwise.

### III. STATISTICAL CLOSURES OF PR-MD

In the micromorphic multiscale decomposition, the fine scale displacement is determined by the following multiplicative decomposition,

$$\mathbf{r}'_{\alpha i} = \mathbf{r}_{\alpha i} - \bar{\mathbf{r}}_{\alpha i}, \quad \mathbf{r}_{\alpha i}(t) := \phi_{\alpha}(t) \cdot \mathbf{S}_i(t), \quad (7)$$

where  $\phi_{\alpha}$  is the micro-deformation tensor of the  $\alpha$ -th cell, and the vector  $\mathbf{S}_i$  belongs to a time-dependent configuration that is overlap with  $\Omega_{\alpha}(0)$ , and  $\mathbf{S}_i(0) = \mathbf{R}_{\alpha i}$ . Since,

$$\dot{\mathbf{r}}_{\alpha i} = \dot{\phi}_{\alpha} \mathbf{S}_i + \phi_{\alpha} \dot{\mathbf{S}}_i \quad (8)$$

It is trivial to show that both

$$\sum_i m_i \mathbf{S}_i = 0, \quad \text{and} \quad \sum_i m_i \dot{\mathbf{S}}_i = 0. \quad (9)$$

For simplicity we only consider the following first-principle Lagrangian for the  $\alpha$ -th cell with pair potential and usual external potential

$$\mathcal{L}_{\alpha} = \frac{1}{2} \sum_i m_i \dot{\mathbf{r}}_i \cdot \dot{\mathbf{r}}_i - \frac{1}{2} \sum_i \sum_{j \neq i} V(r_{ij}) - \sum_i \mathbf{f}_i \cdot \mathbf{r}_i \quad (10)$$

where  $\phi(r_{ij})$  is the pair potential,  $r_{ij} = |\mathbf{r}_{ij}|$  and  $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ ;  $\mathbf{f}_i$  is the external force acting on the atom  $i$ .

By considering the center of mass decomposition, the total kinetic energy becomes,

$$\begin{aligned}
\mathcal{K} &= \frac{1}{2} \sum_i m_i (\dot{\mathbf{r}}_\alpha + \dot{\phi}_\alpha \cdot \mathbf{S}_i + \phi_\alpha \cdot \dot{\mathbf{S}}_i) \\
&\quad \cdot (\dot{\mathbf{r}}_\alpha + \dot{\phi}_\alpha \cdot \mathbf{S}_i + \phi_\alpha \cdot \dot{\mathbf{S}}_i) \\
&= \underbrace{\frac{M}{2} \dot{\mathbf{r}}_\alpha \cdot \dot{\mathbf{r}}_\alpha}_{\mathcal{K}_1} + \underbrace{\frac{1}{2} \sum_i m_i \mathbf{S}_i \cdot (\dot{\phi}_\alpha^T \dot{\phi}_\alpha) \cdot \mathbf{S}_i}_{\mathcal{K}_2} \\
&\quad + \underbrace{\frac{1}{2} \sum_i m_i \dot{\mathbf{S}}_i \cdot \mathbf{C} \cdot \dot{\mathbf{S}}_i}_{\mathcal{K}_3} \\
&\quad + \underbrace{\frac{1}{2} \sum_i m_i (\mathbf{S}_i (\dot{\phi}_\alpha^T \phi_\alpha) \cdot \dot{\mathbf{S}}_i + \dot{\mathbf{S}}_i (\phi_\alpha^T \dot{\phi}_\alpha) \mathbf{S}_i)}_{\mathcal{K}_4}
\end{aligned} \tag{11}$$

The second term of (11) may be written as

$$\mathcal{K}_2 = \frac{1}{2} (\dot{\phi}_\alpha^T \dot{\phi}_\alpha) \sum_i m_i \mathbf{S}_i \otimes \mathbf{S}_i. \tag{12}$$

To link the microscale quantities to macroscale field variables, we introduce the following statistical assumption,

$$\sum_i m_i \mathbf{S}_i \otimes \mathbf{S}_i = I_{IJ} \mathbf{E}_I \otimes \mathbf{E}_J = \text{const}. \tag{13}$$

which we coined as the first Parrinello-Rahman closure condition, and it is obviously valid under equilibrium states.

Let

$$\dot{\phi}_{iI}^\alpha \dot{\phi}_{iJ}^\alpha = \omega_I \omega_J \tag{14}$$

The second term of the kinetic energy may be written as

$$\frac{1}{2} \boldsymbol{\omega} \cdot \mathbf{J} \cdot \boldsymbol{\omega} = \frac{1}{2} J_{IJ} \omega_I \omega_J \tag{15}$$

Note that  $\mathbf{J}$  is not the standard moment inertia tensor. The standard moment inertia tensor is defined as

$$\mathbf{J}_s = \int_V \rho (\|\mathbf{R}\|^2 \mathbf{I}^{(2)} - \mathbf{R} \otimes \mathbf{R}) dV$$

where  $\mathbf{I}^{(2)}$  is the second order unit tensor.

If  $\{\mathbf{E}_I\}$  are principal axes, we have

$$\mathbf{J} = J_{11} \mathbf{E}_1 \otimes \mathbf{E}_1 + J_{22} \mathbf{E}_2 \otimes \mathbf{E}_2 + J_{33} \mathbf{E}_3 \otimes \mathbf{E}_3$$

If  $J_{11} = J_{22} = J_{33} = W$ , we have the following result from Parrinello and Rahman [3],

$$\mathcal{K}_2 = \frac{1}{2} W \text{tr}(\dot{\phi}_\alpha^T \dot{\phi}_\alpha). \tag{16}$$

In practice, one may make the following approximation,

$$\begin{aligned}
\mathbf{J} &= \sum_i m_i \mathbf{S}_i \otimes \mathbf{S}_i \approx \sum_i m_i \mathbf{S}_i(0) \otimes \mathbf{S}_i(0) \\
&= \sum_i m_i \mathbf{R}_{\alpha i} \otimes \mathbf{R}_{\alpha i}
\end{aligned} \tag{17}$$

In [3], Parrinello and Rahman made another choice, and they let

$$\mathcal{K}_4 = \frac{1}{2} \sum_i m_i \left( \mathbf{S}_i (\dot{\phi}_\alpha^T \phi_\alpha) \cdot \dot{\mathbf{S}}_i + \dot{\mathbf{S}}_i (\phi_\alpha^T \dot{\phi}_\alpha) \mathbf{S}_i \right) = 0, \quad (18)$$

which we call as the second Parrinello-Rahman closure.

In this paper, we shall not adopt the second Parrinello-Rahman closure, and instead we shall examine the detailed contribution to the Euler-Lagrange equations from the term  $\mathcal{K}_5$ . First, we may write,

$$\mathcal{K}_4 = \text{tr} \left\{ \dot{\phi}_\alpha \cdot \left( \sum_i m_i \mathbf{S}_i \otimes \dot{\mathbf{S}}_i \right) \cdot \phi_\alpha^T \right\}$$

We can then calculate the following partial derivatives,

$$\frac{\partial \mathcal{K}_4}{\partial \mathbf{S}_i} = \dot{\phi}_\alpha^T \phi_\alpha \cdot \left( \sum_i m_i \dot{\mathbf{S}}_i \right) = 0; \quad (19)$$

$$\frac{\partial \mathcal{K}_4}{\partial \dot{\mathbf{S}}_i} = \phi_\alpha^T \dot{\phi}_\alpha \cdot \left( \sum_i m_i \mathbf{S}_i \right) = 0; \quad (20)$$

$$\frac{\partial \mathcal{K}_4}{\partial \phi_\alpha} = \dot{\phi}_\alpha \cdot \left( \sum_i m_i \mathbf{S}_i \otimes \dot{\mathbf{S}}_i \right) \quad (21)$$

$$\frac{\partial \mathcal{K}_4}{\partial \dot{\phi}_\alpha} = \phi_\alpha \cdot \left( \sum_i m_i \dot{\mathbf{S}}_i \otimes \mathbf{S}_i \right) \quad (22)$$

If we choose the following statistical closures,

$$\left( \sum_i m_i \mathbf{S}_i \otimes \dot{\mathbf{S}}_i \right) = 0, \quad (a)$$

$$\left( \sum_i m_i \dot{\mathbf{S}}_i \otimes \mathbf{S}_i \right) = 0, \quad (b) \quad (23)$$

we shall have  $\frac{\partial \mathcal{K}_4}{\partial \phi_\alpha} = 0$  and  $\frac{\partial \mathcal{K}_4}{\partial \dot{\phi}_\alpha} = 0$ .

In fact, the first Parrinello-Rahman closure implies that

$$\left( \sum_i m_i \mathbf{S}_i \otimes \dot{\mathbf{S}}_i \right) + \left( \sum_i m_i \dot{\mathbf{S}}_i \otimes \mathbf{S}_i \right) = 0, \quad (24)$$

but obviously Eq. (23)(a) and (b) are stronger conditions than (24). Thus the closure conditions (23)(a) and (b) imply the first Parrinello-Rahman closure. In this paper, we call (23)(a) and (b) as the mesoscale closure conditions.

Since  $\mathcal{K}_4$  may be re-written as the following form

$$\mathcal{K}_4 = \sum_i \frac{m_i}{2} \left\{ (\dot{\phi}_\alpha^T \phi_\alpha) : (\mathbf{S}_i \otimes \dot{\mathbf{S}}_i) + (\phi_\alpha^T \dot{\phi}_\alpha) : (\dot{\mathbf{S}}_i \otimes \mathbf{S}_i) \right\}, \quad (25)$$

it is clear that the mesoscale closure can effectively enforce both the first and the second Parrinello-Rahman closures. To justify the above statistical closures, we study the correlation property of the random variables  $\{\mathbf{S}_i\}$ , which have the property,

$$\sum_i \mathbf{S}_i = 0.$$

We first define the following tensorial autocorrelation function, Consider the following tensorial correlation function,

$$\begin{aligned} \mathbf{AC}_1(\tau) &= \frac{1}{2} \left( \langle \mathbf{S}(t) \otimes \mathbf{S}(t+\tau) \rangle + \langle \mathbf{S}(t+\tau) \otimes \mathbf{S}(t) \rangle \right) \\ &:= \frac{1}{2N} \left( \sum_i m_i \mathbf{S}_i(t) \otimes \mathbf{S}_i(t+\tau) + \sum_i m_i \mathbf{S}_i(t+\tau) \otimes \mathbf{S}_i(t) \right) \end{aligned} \quad (26)$$

It is obvious that  $\mathbf{AC}_1(\tau)$  is an even function, and

$$\left. \frac{d}{d\tau} \mathbf{AC}_1 \right|_{\tau=0} = 0, \quad \rightarrow \quad \sum_i m_i \dot{\mathbf{S}}_i(t) \otimes \mathbf{S}_i(t) + \sum_i m_i \mathbf{S}_i(t) \otimes \dot{\mathbf{S}}_i(t) = 0,$$

which is the statement of the first Parrinello-Rahman closure.

Next we consider the following tensorial autocorrelation function,

$$\begin{aligned} \mathbf{AC}_2(\tau) &= \langle \mathbf{S}(t) \otimes \mathbf{S}(t+\tau) \rangle = \frac{1}{2} \left( \langle \mathbf{S}(t) \otimes \mathbf{S}(t+\tau) \rangle + \langle \mathbf{S}(t-\tau) \otimes \mathbf{S}(t) \rangle \right) \\ &:= \frac{1}{2N} \left( \sum_i m_i \mathbf{S}_i(t) \otimes \mathbf{S}_i(t+\tau) + \sum_i m_i \mathbf{S}_i(t-\tau) \otimes \mathbf{S}_i(t) \right) \end{aligned} \quad (27)$$

If we assume that the autocorrelation is a constant or slowly changing tensor in the neighborhood of  $\tau = 0$ , we can then have

$$\left. \frac{d}{d\tau} \mathbf{AC}_2 \right|_{\tau=0} = 0 \quad \rightarrow \quad \sum_i m_i \dot{\mathbf{S}}_i(t) \otimes \mathbf{S}_i(t) = \sum_i m_i \mathbf{S}_i(t) \otimes \dot{\mathbf{S}}_i(t) = 0, \quad (28)$$

which is the mesoscale closure.

#### IV. ANDERSEN-PARRINELLO-RAHMAN MOLECULAR DYNAMICS

As Ray and Rahman [15] referred it as  $N\sigma H$  ensemble MD, the anisotropic Lagrangian of the Andersen-Parrinello-Rahman molecular dynamics for an isoenthalpic-isobaric ensemble is,

$$\begin{aligned} \mathcal{L}_\alpha &= \frac{1}{2} \sum_i m_i \mathbf{S}_i \cdot (\dot{\phi}_\alpha^T \dot{\phi}_\alpha) \cdot \mathbf{S}_i \\ &+ \frac{1}{2} \sum_i m_i \dot{\mathbf{S}}_i \cdot \mathbf{C} \cdot \dot{\mathbf{S}}_i \\ &- \frac{1}{2} \sum_i \sum_{j \neq i} V(r_{ij}) - \sum_i \mathbf{f}_i \cdot \mathbf{r}_i - \tilde{\mathbf{P}} : \phi_\alpha \Omega_\alpha \end{aligned} \quad (29)$$

where  $\tilde{\mathbf{P}}$  is the thermodynamic conjugate stress measure of  $\phi_\alpha$ .

The basic equations of the PR molecular dynamics are

$$\frac{d}{dt} \frac{\partial \mathcal{L}_\alpha}{\partial \dot{\phi}_\alpha} - \frac{\partial \mathcal{L}_\alpha}{\partial \phi_\alpha} = 0, \quad (30)$$

$$\frac{d}{dt} \frac{\partial \mathcal{L}_\alpha}{\partial \dot{\mathbf{S}}_i} - \frac{\partial \mathcal{L}_\alpha}{\partial \mathbf{S}_i} = 0. \quad (31)$$

Following the standard procedure, one may derive the equations of motions for the multiscale molecular dynamics in the  $\alpha$ -th cell,

$$\ddot{\mathbf{S}}_i = - \sum_{j \neq i} \left( \frac{V'(r_{ij})}{m_i r_{ij}} \right) (\mathbf{S}_i - \mathbf{S}_j) - \mathbf{C}^{-1} \cdot \dot{\mathbf{C}} \cdot \dot{\mathbf{S}}_i - \phi_\alpha^{-1} \cdot \mathbf{f}_i \otimes \mathbf{S}_i, \text{ and} \quad (32)$$

$$\ddot{\phi}_\alpha \cdot \mathbf{J} = J \sigma_\alpha \phi_\alpha^{-T} \Omega_0 - \tilde{\mathbf{P}} \Omega_{\alpha 0} - \sum_i \mathbf{f}_i \otimes \mathbf{S}_i \quad (33)$$

where  $\sigma_\alpha$  is the Virial stress defined as

$$\sigma_\alpha = \frac{1}{\Omega_\alpha} \sum_i \left( -m_i \mathbf{v}'_i \otimes \mathbf{v}'_i + \sum_{j \neq i} V'(r_{ij}) \left( \frac{\mathbf{S}_{ij} \otimes \mathbf{S}_{ij}}{r_{ij}} \right) \right) \quad (34)$$

where  $\Omega_\alpha(t)$  is the volume of the  $\alpha$ -th MD cell, and the fine scale velocity is defined as

$$\mathbf{v}'_i = \phi_\alpha \cdot \dot{\mathbf{S}}_i \quad (35)$$

One can verify that indeed

$$\sum_i m_i \mathbf{v}'_i = 0 .$$

That is  $\mathbf{v}'_i$  is a peculiar velocity, but  $\mathbf{v}'_i \neq \dot{\mathbf{r}}_{\alpha i}$ .

Based on nonlinear continuum mechanics [16], one may define the first Piola-Kirchhoff stress as,

$$\mathbf{P}_\alpha = J \boldsymbol{\sigma}_\alpha \phi_\alpha^{-1} \quad (36)$$

so the third equation becomes

$$\ddot{\phi}_\alpha \cdot \mathbf{J} = (\mathbf{P}_\alpha - \tilde{\mathbf{P}}) \Omega_{\alpha 0} - \sum_i \mathbf{f}_i \otimes \mathbf{S}_i \quad (37)$$

In above derivations, Eq. (32)-(33) are essentially the same as those of Parrinello and Rahman's original formulation except the external potential energy. To this end, we have present a first-principle based justification of APR molecular dynamics.

For APR-MD, in the original single cell of atom ensemble, the position of the center of mass is fixed, because the periodic boundary condition is used. Hence

$$\frac{\partial}{\partial \mathbf{r}_\alpha} V(r_{ij}) = 0 .$$

In this case, the centers of mass of different cells only undergo rigid motion.

In coupling of multiple different cells without imposing the periodic boundary condition, the relative displacements of each center of mass will be different, so that

$$\frac{\partial}{\partial \mathbf{r}_\alpha} V(r_{ij}) \neq 0 .$$

In the next Sections, we shall discuss the multiscale micromorphic molecular dynamics that is applicable to arbitrary domain with arbitrary boundary conditions.

## V. MULTISCALE MICROMORPHIC MOLECULAR DYNAMICS

In this Section, we shall extend the periodic boundary condition based APR-MD to an arbitrary finite size multiscale micromorphic molecular dynamics.

To extend APR molecular dynamics to mesoscale scale and continuum scale with arbitrary boundary conditions, we propose the following three scale kinematic decomposition,

$$\mathbf{r}_i = \mathbf{r}_\alpha + \phi_\alpha \cdot \mathbf{S}_i \quad (38)$$

$$\phi_\alpha = \boldsymbol{\chi}_\alpha \cdot \mathbf{F}_\alpha \quad (39)$$

where  $\mathbf{r}_i$  is the position of the  $i$ -th atom of the system in the deformed configuration;  $\mathbf{S}_i$  the random vibration position of the  $i$ -th atom in the referential configuration;  $\mathbf{r}_\alpha$  is the center of mass of  $\alpha$ -th unit cell;  $\boldsymbol{\chi}_\alpha$  is the micro deformation of the  $\alpha$ -th unit cell, and  $\mathbf{F}_\alpha$  is the coarse scale deformation gradient, which is determined by the relative position of the centers of mass of different cells. The three independent kinematic variables at three different scales are:  $\{\mathbf{S}_i, \boldsymbol{\chi}_\alpha$ , and  $\mathbf{r}_\alpha\}$ . The novelty of the proposed multiscale decomposition is the multiplicative multiscale decomposition Eq. (39). We note that even though the coarse scale deformation gradient only depends on the relative position of the centers of mass of different cells, i.e.  $\mathbf{F}_\alpha = \mathbf{F}_\alpha(\{\mathbf{r}_\beta\})$ , it may take different values for different atoms in a same cell, i.e.  $\mathbf{F}_\alpha(\mathbf{R}_i, \{\mathbf{r}_\beta\}) \neq \mathbf{F}_\alpha(\mathbf{R}_j, \{\mathbf{r}_\beta\})$ ,  $\mathbf{R}_i, \mathbf{R}_j \in \Omega_{\alpha 0}$ . More precisely, Eqs. (38) and (39) may be written as

$$\mathbf{r}_i = \mathbf{r}_\alpha + \phi_\alpha(\mathbf{R}_i) \cdot \mathbf{S}_i \quad (40)$$

$$\phi_\alpha(\mathbf{R}_i) = \boldsymbol{\chi}_\alpha \cdot \mathbf{F}_\alpha(\mathbf{R}_i, \{\mathbf{r}_\beta\}) \quad (41)$$

with the understanding that  $\mathbf{R}_i$  is just an interpolation variable, and  $\mathbf{F}_\alpha = \mathbf{F}_\alpha(\mathbf{R}_i)$  does not depend on dynamics variable  $\mathbf{S}_i$ . In the subsequent Sections, we shall discuss the difference between the case that  $\mathbf{F}_\alpha$  depends on  $\mathbf{R}_i$  in a

fixed cell and the case that  $\mathbf{F}_\alpha$  is a constant tensor in the entire cell. If we denote the lattice spacing as  $\ell_a$ , the unit cell size as  $\ell_c$ , and the nonlocal support of a center of mass particle as  $\ell_r$ , we have

$$\ell_a < \ell_c < \ell_r .$$

The time derivatives of the independent kinematic variables are,

$$\begin{aligned}\dot{\mathbf{r}}_\alpha &= \dot{\mathbf{r}}_\alpha + \dot{\phi}_\alpha \cdot \mathbf{S}_i + \phi_\alpha \cdot \dot{\mathbf{S}}_i \\ \dot{\phi}_\alpha &= \dot{\chi}_\alpha \cdot \mathbf{F}_\alpha + \chi_\alpha \cdot \dot{\mathbf{F}}_\alpha\end{aligned}$$

If we denote the time scale of  $\dot{\mathbf{S}}_i$  as  $t_s$ , the time scale of  $\dot{\chi}_\alpha$  as  $t_c$ , and time scale  $\dot{\mathbf{r}}_\alpha$  as  $t_r$ , we again have

$$t_s < t_c < t_r .$$

We start with a first principle Lagrange (10) of a multiscale ensemble (multiple cells) system in terms of following multiscale decomposition,

$$\begin{aligned}\mathcal{L}_m &= \frac{1}{2} \sum_{\beta} M_{\beta} \dot{\mathbf{r}}_{\beta} \cdot \dot{\mathbf{r}}_{\beta} + \frac{1}{2} \sum_{\beta} \mathbf{J}_{\beta} : (\dot{\phi}_{\beta}^T \dot{\phi}_{\beta}) \\ &+ \frac{1}{2} \sum_{\beta} \sum_i m_i \dot{\mathbf{S}}_i \cdot \mathbf{C}_{\beta} \cdot \dot{\mathbf{S}}_i - \frac{1}{2} \sum_{\beta} \sum_{\gamma} \sum_{i \in \beta, j \in \gamma} V(r_{ij}) \\ &- \sum_{\beta} \sum_{i \in \beta} \mathbf{f}_i \cdot \mathbf{r}_i\end{aligned}\tag{42}$$

where  $\beta, \gamma$  are cell indices, and the abbreviation  $i \in \beta$  means that the  $i$ -th atom belongs to the  $\beta$ -th cell.  $\mathbf{C}_{\beta} := \phi_{\beta}^T \phi_{\beta}$  is the micro right Cauchy-Green tensor for total deformation. We denote that  $M_{\beta} = \sum_{i \in \beta} m_i$  and  $\mathbf{B}_{\beta} = \sum_{i \in \beta} \mathbf{f}_i$ . In the rest of this paper, we always assume that the Roman index  $i$  is used to make the atoms inside the  $\beta$ -th cell, whereas the Roman index  $j$  is designated to denote the atoms inside the  $\gamma$ -th cell.

For simplicity, we choose three independent field variables,  $\mathbf{r}_\alpha$ ,  $\phi_\alpha$ , and  $\mathbf{S}_i$  for three scales, i.e.  $\mathcal{L}_m = \mathcal{L}_m(\mathbf{r}_\alpha, \phi_\alpha, \mathbf{S}_i)$ . We postulate the following principle of multiscale stationary action,

$$\delta S[\mathbf{r}_\alpha, \phi_\alpha, \{\mathbf{S}_i\}] = \int_{t_1}^{t_2} \delta \mathcal{L}_m(\dot{\mathbf{r}}_\alpha, \dot{\phi}_\alpha, \{\dot{\mathbf{S}}_i\}, \mathbf{r}_\alpha, \phi_\alpha, \{\mathbf{S}_i\}, t) dt = 0 .$$

The Lagrangian equations of the multiscale micromorphic molecular dynamics can be derived as follows,

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{r}}_\alpha} - \frac{\partial \mathcal{L}_m}{\partial \mathbf{r}_\alpha} = 0,\tag{43}$$

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\alpha} - \frac{\partial \mathcal{L}_m}{\partial \phi_\alpha} = 0,\tag{44}$$

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{S}}_i} - \frac{\partial \mathcal{L}_m}{\partial \mathbf{S}_i} = 0 .\tag{45}$$

Note that one may choose  $\phi_\alpha$  instead of  $\chi_\alpha$  as the independent field variable for mesoscale dynamic equations. Since  $\phi_\alpha = \chi_\alpha \cdot \mathbf{F}_\alpha$ , the two choices will be equivalent.

### A. Coarse scale dynamic equations

We start by deriving some useful relations that are needed in the subsequent derivations. Since we know that

$$\mathbf{F}_\beta = \mathbf{F}_\beta(\{\mathbf{r}_\alpha\}), \quad \dot{\mathbf{F}}_\beta = \dot{\mathbf{F}}_\beta(\{\mathbf{r}_\alpha\}, \{\dot{\mathbf{r}}_\alpha\})\tag{46}$$

then we have

$$\dot{\mathbf{F}}_\beta = \sum_{\alpha} \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} \dot{\mathbf{r}}_\alpha,\tag{47}$$



which leads to the relation,

$$\frac{\partial \dot{\mathbf{F}}_\beta}{\partial \dot{\mathbf{r}}_\alpha} = \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha}, \quad (48)$$

and

$$\ddot{\mathbf{F}}_\beta = \sum_\alpha \left( \frac{d}{dt} \left( \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} \right) \dot{\mathbf{r}}_\alpha + \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} \ddot{\mathbf{r}}_\alpha \right). \quad (49)$$

On the other hand, we may derive,

$$\ddot{\mathbf{F}}_\beta = \sum_\alpha \left( \frac{\partial \dot{\mathbf{F}}_\beta}{\partial \mathbf{r}_\alpha} \dot{\mathbf{r}}_\alpha + \frac{\partial \dot{\mathbf{F}}_\beta}{\partial \dot{\mathbf{r}}_\alpha} \ddot{\mathbf{r}}_\alpha \right) \quad (50)$$

Comparing equations (49) and (50) and utilizing (48), we obtain

$$\frac{d}{dt} \left( \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} \right) = \frac{\partial \dot{\mathbf{F}}_\beta}{\partial \mathbf{r}_\alpha}. \quad (51)$$

Next we are first taking time derivative on  $\phi_\beta$ ,

$$\dot{\phi}_\beta = \dot{\chi}_\beta \mathbf{F}_\beta + \chi_\beta \dot{\mathbf{F}}_\beta, \quad (52)$$

and then we can find that

$$\frac{\partial \dot{\phi}_\beta}{\partial \dot{\mathbf{r}}_\alpha} = \chi_\beta \frac{\partial \dot{\mathbf{F}}_\beta}{\partial \dot{\mathbf{r}}_\alpha} = \chi_\beta \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} = \frac{\partial \phi_\beta}{\partial \mathbf{r}_\alpha}. \quad (53)$$

By virtue of Eqs. (51) ~ (53), we have

$$\begin{aligned} \frac{\partial \dot{\phi}_\beta}{\partial \mathbf{r}_\alpha} &= \dot{\chi}_\beta \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} + \chi_\beta \frac{\partial \dot{\mathbf{F}}_\beta}{\partial \mathbf{r}_\alpha} \\ &= \dot{\chi}_\beta \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} + \chi_\beta \frac{d}{dt} \left( \frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} \right) \\ &= \frac{d}{dt} \left( \frac{\partial \phi_\beta}{\partial \mathbf{r}_\alpha} \right) = \frac{d}{dt} \left( \frac{\partial \dot{\phi}_\beta}{\partial \dot{\mathbf{r}}_\alpha} \right). \end{aligned} \quad (54)$$

This relation is needed in the subsequent derivation.

Reconsidering the Lagrangian equation at the coarse scale and utilizing the above relation, we have

$$\begin{aligned} \frac{d}{dt} \left( \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{r}}_\alpha} \right) &= \frac{d}{dt} \left( \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{r}}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \cdot \frac{\partial \dot{\phi}_\beta}{\partial \dot{\mathbf{r}}_\alpha} \right) \\ &= M_\alpha \ddot{\mathbf{r}}_\alpha + \sum_\beta \frac{d}{dt} \left( \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \right) \cdot \frac{\partial \dot{\phi}_\beta}{\partial \dot{\mathbf{r}}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \cdot \frac{d}{dt} \left( \frac{\partial \dot{\phi}_\beta}{\partial \dot{\mathbf{r}}_\alpha} \right) \\ &= M_\alpha \ddot{\mathbf{r}}_\alpha + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \phi_\beta} \cdot \frac{\partial \phi_\beta}{\partial \mathbf{r}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \cdot \frac{\partial \dot{\phi}_\beta}{\partial \mathbf{r}_\alpha} \end{aligned} \quad (55)$$

On the other hand,

$$\begin{aligned} \frac{\partial \mathcal{L}_m}{\partial \mathbf{r}_\alpha} &= \frac{\partial \mathcal{L}_m}{\partial \mathbf{r}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \phi_\beta} \cdot \frac{\partial \phi_\beta}{\partial \mathbf{r}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \cdot \frac{\partial \dot{\phi}_\beta}{\partial \mathbf{r}_\alpha} \\ &= - \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} V'(r_{ij}) \frac{\mathbf{r}_{ij}}{|\mathbf{r}_{ij}|} - \sum_{i \in \alpha} \mathbf{f}_i + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \phi_\beta} \cdot \frac{\partial \phi_\beta}{\partial \mathbf{r}_\alpha} + \sum_\beta \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\beta} \cdot \frac{\partial \dot{\phi}_\beta}{\partial \mathbf{r}_\alpha}. \end{aligned} \quad (56)$$

Combining Eqs. (55) and (56) and utilizing the coarse scale Lagrangian equation,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{r}}_\alpha} \right) - \frac{\partial \mathcal{L}_m}{\partial \mathbf{r}_\alpha} = 0 ,$$

we finally have

$$M_\alpha \ddot{\mathbf{r}}_\alpha + \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} V'(r_{ij}) \frac{\mathbf{r}_{ij}}{|\mathbf{r}_{ij}|} + \mathcal{B}_\alpha = \mathbf{0} . \quad (57)$$

It is clear that the first term is the coarse scale inertia term, and the second term is the cell-cell interaction force, and the third term is the external force acting on the center of mass of the  $\alpha$ -th cell.

## B. Mesoscale dynamic equations

Second, we exam the mesoscale Lagrangian equation,

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\alpha} - \frac{\partial \mathcal{L}_m}{\partial \phi_\alpha} = 0 .$$

For a systematic derivation, we denote

$$\begin{aligned} \mathbf{r}_i &= \mathbf{r}_\beta + \phi_\alpha \cdot \mathbf{S}_i, \quad \text{and} \quad \mathbf{r}_j = \mathbf{r}_\gamma + \phi_\gamma \cdot \mathbf{S}_j \rightarrow \\ \mathbf{r}_{ij} &= \mathbf{r}_j - \mathbf{r}_i = \mathbf{r}_{\beta\gamma} + \phi_\gamma \cdot \mathbf{S}_j - \phi_\beta \cdot \mathbf{S}_i \end{aligned} \quad (58)$$

where

$$\phi_\beta = \chi_\beta \cdot \mathbf{F}_\beta \quad \text{and} \quad \phi_\gamma = \chi_\gamma \cdot \mathbf{F}_\gamma$$

To facilitate the subsequent derivation, we first consider the derivative terms with respect to the chosen mesoscale variable, i.e.  $\dot{\phi}_\alpha$  and  $\phi_\alpha$  :

1.

$$\frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\alpha} = \dot{\phi}_\alpha \cdot \mathbf{J}_\alpha$$

where  $\mathbf{J}_\alpha = \sum_i m_i \mathbf{S}_i \otimes \mathbf{S}_i \approx \sum_i m_i \mathbf{R}_i \otimes \mathbf{R}_i$ . Hence

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_m}{\partial \dot{\phi}_\alpha} \right) = \frac{d}{dt} (\dot{\phi}_\alpha \cdot \mathbf{J}_\alpha) = \ddot{\phi}_\alpha \cdot \mathbf{J}_\alpha$$

2.

$$\frac{\partial \mathcal{C}_\alpha}{\partial \phi_\alpha} = 2\phi_\alpha$$

3.

$$(a) \beta = \gamma : \quad \frac{\partial r_{ij}}{\partial \phi_\alpha} = \frac{\mathbf{r}_{ij}}{r_{ij}} \cdot \frac{\partial \mathbf{r}_{ij}}{\partial \phi_\alpha} = \left( \frac{\mathbf{r}_{ij}}{r_{ij}} \otimes \mathbf{S}_{ij} \right) \delta_{\alpha\beta} \quad (59)$$

$$(b) \beta \neq \gamma : \quad \frac{\partial r_{ij}}{\partial \phi_\alpha} = \frac{\mathbf{r}_{ij}}{r_{ij}} \cdot \frac{\partial \mathbf{r}_{ij}}{\partial \phi_\alpha} = \frac{\mathbf{r}_{ij}}{r_{ij}} \otimes (\delta_{\alpha\gamma} \mathbf{S}_j - \delta_{\alpha\beta} \mathbf{S}_i) . \quad (60)$$

Hence,

$$\begin{aligned} (a) \beta = \gamma : \quad \frac{\partial \mathcal{L}_m}{\partial \phi_\alpha} &= \frac{1}{2} \sum_i m_i \dot{\mathbf{S}}_i \frac{\partial \mathcal{C}_\alpha}{\partial \phi_\alpha} \dot{\mathbf{S}}_i - \frac{1}{2} \frac{\partial}{\partial \phi_\alpha} \sum_\beta \sum_{j \neq i, i, j \in \beta} V(r_{ij}) \\ &= \phi_\alpha \cdot \sum_i m_i \dot{\mathbf{S}}_i \otimes \dot{\mathbf{S}}_i - \frac{1}{2} \sum_{j \neq i, i, j \in \alpha} \frac{V'(r_{ij})}{r_{ij}} \phi_\alpha \cdot \mathbf{S}_{ij} \otimes \mathbf{S}_{ij} \\ (b) \beta \neq \gamma : \quad \frac{\partial \mathcal{L}_m}{\partial \phi_\alpha} &= \frac{1}{2} \sum_i m_i \dot{\mathbf{S}}_i \frac{\partial \mathcal{C}_\alpha}{\partial \phi_\alpha} \dot{\mathbf{S}}_i - \frac{1}{2} \frac{\partial}{\partial \phi_\alpha} \sum_\beta \sum_\gamma \sum_{i \in \beta, j \in \gamma} V(r_{ij}) \\ &= \phi_\alpha \cdot \sum_i m_i \dot{\mathbf{S}}_i \otimes \dot{\mathbf{S}}_i - \frac{1}{2} \sum_\beta \sum_\gamma \sum_{i \in \beta, j \in \gamma} \frac{V'(r_{ij})}{r_{ij}} \mathbf{r}_{ij} \otimes (\delta_{\alpha\gamma} \mathbf{S}_j - \delta_{\alpha\beta} \mathbf{S}_i) \end{aligned}$$

The dynamic equations at mesoscale have the form,

$$\begin{aligned} & \ddot{\phi}_\alpha \cdot \mathbf{J}_\alpha - \phi_\alpha \sum_i m_i \dot{\mathbf{S}}_i \otimes \dot{\mathbf{S}}_i + \sum_{i,j \in \alpha, i \neq j} \frac{V'(r_{ij})}{r_{ij}} \phi_\alpha \cdot \mathbf{S}_i \otimes \mathbf{S}_i \\ & + \sum_{\beta \neq \gamma} \sum_{i \in \beta, j \in \gamma} \frac{V'(r_{ij})}{r_{ij}} \mathbf{r}_{ij} \otimes (\delta_{\alpha\gamma} \mathbf{S}_j - \delta_{\alpha\beta} \mathbf{S}_i) + \sum_{i \in \alpha} \mathbf{f}_i \otimes \mathbf{S}_i = 0. \end{aligned} \quad (61)$$

where  $\mathbf{r}_{ij} = \mathbf{r}_{\beta\gamma} + \phi_\gamma \cdot \mathbf{S}_j - \phi_\beta \cdot \mathbf{S}_i$ .

Define the mesoscale  $2^{nd}$  Piola-Kirchhoff stress tensor,

$$\mathcal{S}_\alpha^{int} := \frac{1}{\Omega_{\alpha 0}} \sum_{i \in \alpha} \left( -m_i \dot{\mathbf{S}}_i \otimes \dot{\mathbf{S}}_i + \sum_{j \in \alpha, j \neq i} \frac{V'(r_{ij})}{r_{ij}} \mathbf{S}_i \otimes \mathbf{S}_i \right) \quad (62)$$

$$\mathcal{S}_\alpha^{ext} = \frac{1}{\Omega_{\alpha 0}} \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} \frac{V'(r_{ij})}{r_{ij}} \mathbf{r}_{ij} \otimes \mathbf{S}_i \quad (63)$$

where  $\mathbf{r}_{ij} = \mathbf{r}_{\alpha\beta} + \phi_\beta \cdot \mathbf{S}_j - \phi_\alpha \cdot \mathbf{S}_i$ .

The mesoscale dynamics equations can be recast into

$$\ddot{\phi}_\alpha \cdot \mathbf{J}_\alpha + \phi_\alpha \cdot (\mathcal{S}_\alpha^{int} - \mathcal{S}_\alpha^{ext}) \Omega_{\alpha 0} + \mathbf{M}_\alpha = 0,$$

where  $\mathbf{M}_\alpha = \sum_{i \in \alpha} \mathbf{f}_i \otimes \mathbf{S}_i$  is the mesoscale external couple. Note that Eqs. (62) and (63) are insightful, because it

resolves one of outstanding debates on the definition of the Virial Stress. Eq. (62) is basically the mathematical definition of the Virial stress e.g. [17, 18]. However, Zhu [19] argued that the kinetic energy part should be dropped out in the stress calculation, even though many disagreed, e.g. [20, 21]. We now see from Eqs. (62) and (63) that if the stress is internally generated, the definition of the virial stress is the original definition of the virial stress; but if the stress is an external stress, then the kinetic energy part should drop out from its expression. This is because that the current formulation of the multiscale micromorphic molecular dynamics is formulated under adiabatic condition, which does not consider the heat exchange among the cells.

### C. Microscale dynamic equations

For simplicity, we re-index the multiscale Lagrangian as

$$\begin{aligned} \mathcal{L}_m &= \sum_\alpha \frac{M_\alpha}{2} \dot{\mathbf{r}}_\alpha \cdot \dot{\mathbf{r}}_\alpha + \frac{1}{2} \sum_\alpha \mathbf{J}_\alpha : (\dot{\phi}_\alpha^T \dot{\phi}_\alpha) \\ &+ \frac{1}{2} \sum_\alpha \sum_i m_i \dot{\mathbf{S}}_i \cdot \mathbf{C}_\alpha \cdot \dot{\mathbf{S}}_i - \frac{1}{2} \sum_\alpha \sum_\beta \sum_{i \neq j} V(r_{ij}) \\ &- \sum_\alpha \sum_i \mathbf{f}_i \cdot \phi_\alpha \cdot \mathbf{S}_i - \sum_\alpha \mathcal{B}_\alpha \cdot \mathbf{r}_\alpha \end{aligned} \quad (64)$$

where the microscale variable  $\mathbf{S}_i, i \in \alpha$  and  $\mathbf{S}_j, j \in \beta$ .

$$(a) \alpha = \beta, \quad \mathbf{r}_{ij} = \phi_\alpha \cdot \mathbf{S}_{ij}, \quad \frac{\partial r_{ij}}{\partial \mathbf{S}_i} = -\frac{\mathbf{r}_{ij}}{r_{ij}} \cdot \phi_\alpha = -\frac{\mathbf{C}_\alpha \cdot \mathbf{S}_{ij}}{r_{ij}} \quad (65)$$

$$(b) \alpha \neq \beta: \quad \mathbf{r}_{ij} = \mathbf{r}_{\alpha\beta} + (\phi_\beta \cdot \mathbf{S}_j - \phi_\alpha \cdot \mathbf{S}_i), \quad \frac{\partial r_{ij}}{\partial \mathbf{S}_i} = -\frac{\mathbf{r}_{ij}}{r_{ij}} \cdot \phi_\alpha. \quad (66)$$

Evaluating the fine scale Lagrangian equation for  $i \in \alpha$ ,

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{S}}_i} - \frac{\partial \mathcal{L}_m}{\partial \mathbf{S}_i} = 0, \quad i \in \alpha$$

we have

$$\frac{d}{dt} \frac{\partial \mathcal{L}_m}{\partial \dot{\mathbf{S}}_i} = m_i (\mathbf{C}_\alpha \ddot{\mathbf{S}}_i + \dot{\mathbf{C}}_\alpha \cdot \dot{\mathbf{S}}_i)$$

and

$$\begin{aligned} (a) \ \alpha = \beta : \quad \frac{\partial \mathcal{L}_m}{\partial \mathbf{S}_i} &= -\frac{1}{2} \sum_{j \neq i} \left( \frac{V'(r_{ij})}{r_{ij}} \mathbf{C}_\alpha \cdot \mathbf{S}_{ij} \right) \\ (b) \ \alpha \neq \beta : \quad \frac{\partial \mathcal{L}_m}{\partial \mathbf{S}_i} &= -\frac{1}{2} \sum_{\alpha \neq \beta} \sum_{j \neq i} \left( \frac{V'(r_{ij})}{r_{ij}} \phi_\alpha^T \cdot \mathbf{r}_{ij} \right) \end{aligned} \quad (67)$$

where  $\mathbf{r}_{ij} = \mathbf{r}_{\alpha\beta} + \phi_\beta \cdot \mathbf{S}_j - \phi_\alpha \cdot \mathbf{S}_i$ .

Finally, we can express the fine scale dynamics equations as,

$$(a) \ \alpha = \beta : \quad \ddot{\mathbf{S}}_i = -\frac{1}{2} \sum_{j \neq i} \left( \frac{V'(r_{ij})}{r_{ij}} \mathbf{S}_{ij} \right) - \mathbf{C}_\alpha^{-1} \dot{\mathbf{C}}_\alpha \cdot \dot{\mathbf{S}}_i \quad (68)$$

$$(b) \ \alpha \neq \beta : \quad \ddot{\mathbf{S}}_i = -\frac{1}{2} \phi_\alpha^{-1} \sum_{\alpha \neq \beta} \sum_{j \neq i} \left( \frac{V'(r_{ij})}{r_{ij}} (\mathbf{r}_{\alpha\beta} + \phi_\beta \cdot \mathbf{S}_j - \phi_\alpha \cdot \mathbf{S}_i) \right) - \mathbf{C}_\alpha^{-1} \dot{\mathbf{C}}_\alpha \cdot \dot{\mathbf{S}}_i \quad (69)$$

Combining the two equations, we finally have

$$\ddot{\mathbf{S}}_i + \frac{1}{2} \phi_\alpha^{-1} \sum_{\beta} \sum_{i \neq j} \left( \frac{V'(r_{ij})}{r_{ij}} (\mathbf{r}_{\alpha\beta} + \phi_\beta \cdot \mathbf{S}_j - \phi_\alpha \cdot \mathbf{S}_i) \right) + \mathbf{C}_\alpha^{-1} \dot{\mathbf{C}}_\alpha \cdot \dot{\mathbf{S}}_i + \phi_\alpha^{-1} \cdot \mathbf{f}_i = 0. \quad (70)$$

where  $i \in \alpha$ .

## VI. COARSE GRAINED MOLECULAR DYNAMICS

By now, we have derived the exact and complete governing equations for a three-scale micromorphic molecular dynamics, which are based on the first principle Lagrangian. This novel multiscale structure is an intrinsic property of the original molecular dynamics. The only extrinsic parameter is the size of the cell.

As one can find that the motions each scale are strongly coupled to the others. Thus we are able to couple them seamlessly. On the other hand, different from most of the multiscale methods that have been developed in recent years, whose main purpose and advantages are the reduction of computation cost, the proposed MMMD method is a different multiscale paradigm. First, the MMMD is actually more complex and expensive than the original MD, because we have to three sets of equations in three different scales concurrently, and the time integration or the time scale for three sets of dynamics equations are the same. However, the expense of this complexity will allow us to use MD as a nanomechanics tool to simulate finite size problems with arbitrary boundary conditions.

Moreover, the multiscale micromorphic molecular dynamics discovered in this work provides the theoretical foundation for us to derive or to construct the coarse-grained molecular dynamics. For instance, we can shut off molecular dynamics in one or two scales to perform a single scale fast computation. To illustrate this point, we demonstrate in the following how to construct a coarse-scale molecular dynamics.

We first propose to adopt the Reproducing Kernel Particle Method [22] or the state-based Peridynamics [23] techniques in construct the discrete deformation gradient.

To define the coarse scale representation, we choose the coordinates of the center of mass of the each cell as the coarse scale degrees of freedom, so that the first principle Lagrange (10) is the multiscale Lagrange without the need of further modification. However, since the independent variable in the coarse scale is the position of center of mass,  $\mathbf{r}_\alpha$ , and we must link the coarse scale deformation gradient with  $\mathbf{r}_\alpha$ . This can be done by employing an approach adopting by the reproducing kernel particle method or the state-based peridynamics [23], in which the discrete deformation gradient is constructed as,

$$\mathbf{F}_\alpha = \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{r}_{\alpha\beta} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1} \quad (71)$$

where  $\mathbf{R}_{\alpha\beta} := \mathbf{R}_\beta - \mathbf{R}_\alpha$ ;  $\mathbf{r}_{\alpha\beta} = \mathbf{r}_\beta - \mathbf{r}_\alpha$ , and

$$\mathbf{K}_\alpha := \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{R}_{\alpha\beta} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \quad (72)$$

and it is called as the moment function, which is a second order tensor. Note that in Eqs. (71) and (72),  $\omega(|\mathbf{R}_{\alpha i}|)$  is a localized window function, and the common choices are the Gaussian function or the cubic spline function.

The Gaussian is defined as

$$\omega_h(\mathbf{x}) = \frac{1}{(\pi h^2)^{d/2}} \exp\left(-\frac{\mathbf{x} \cdot \mathbf{x}}{h^2}\right) \quad (73)$$

The following cubic spline function is also often chosen in the computation,

$$\omega_h(q) = \frac{A}{h^d} \begin{cases} 1 - \frac{3}{2}q^2 + \frac{3}{4}q^3, & 0 \leq q < 1 \\ \frac{1}{4}(2-q)^3, & 1 \leq q \leq 2 \\ 0, & \text{otherwise} \end{cases} \quad (74)$$

where  $d$  is number of space dimension,  $h$  is the support size, and

$$A = \begin{cases} 2/3 & 1d \\ 10/(2\pi) & 2d \\ 1/\pi & 3d \end{cases}$$

If we assume that the Cauchy-Born rule may be applied for the coarse scale displacement field, i.e.

$$\mathbf{r}_{\alpha\beta} = \mathbf{F}_\alpha \mathbf{R}_{\alpha\beta}. \quad (75)$$

By substituting (75) into (71), we can obtain,

$$\begin{aligned} \mathbf{F}_\alpha &= \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{r}_{\alpha\beta} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1} \\ &= \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{F}_\alpha \mathbf{R}_{\alpha\beta} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1} \\ &= \mathbf{F}_\alpha \end{aligned}$$

We can use all the formulas that are derived in the last Section without any modification, except that we need to explicitly evaluate,

$$\frac{\partial \mathbf{F}_\alpha}{\partial \mathbf{r}_\alpha} = - \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{I}^{(2)} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1} = -\mathbf{I}^{(2)} \otimes \mathbf{D}_\alpha, \quad \beta \neq \alpha, \quad (76)$$

where

$$\mathbf{D}_\alpha = \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1}$$

is a vector.

The same is true for the time derivative of deformation gradient, i.e.

$$\frac{\partial \dot{\mathbf{F}}_\alpha}{\partial \dot{\mathbf{r}}_\alpha} = - \left( \sum_{\beta=1}^{N_h} \omega(|\mathbf{R}_{\alpha\beta}|) \mathbf{I}^{(2)} \otimes \mathbf{R}_{\alpha\beta} \Delta V_\beta \right) \cdot \mathbf{K}_\alpha^{-1} = -\mathbf{I}^{(2)} \otimes \mathbf{D}_\alpha, \quad \beta \neq \alpha. \quad (77)$$

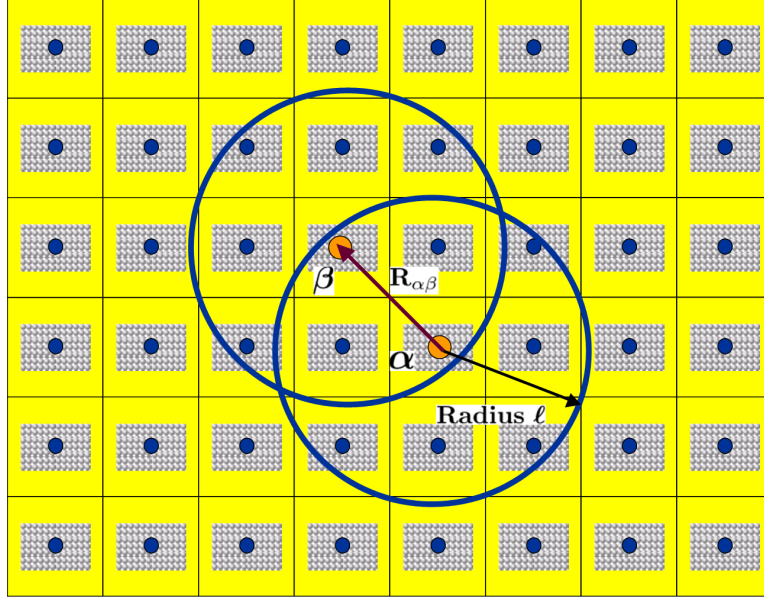


FIG. 1: Nonlocal multiscale micromorphic particle dynamics

In general,

$$\frac{\partial \mathbf{F}_\beta}{\partial \mathbf{r}_\alpha} = \left( \sum_{\gamma=1}^{N_h} \omega(|\mathbf{R}_{\beta\gamma}|) (\delta_{\alpha\gamma} - \delta_{\alpha\beta}) \mathbf{I}^{(2)} \otimes \mathbf{R}_{\beta\gamma} \Delta V_\gamma \right) \cdot \mathbf{K}_\beta^{-1} = -\mathbf{I}^{(2)} \otimes \mathbf{D}_\beta, \quad \gamma \neq \beta, \quad (78)$$

and

$$\frac{\partial \dot{\mathbf{F}}_\beta}{\partial \dot{\mathbf{r}}_\alpha} = \left( \sum_{\gamma=1}^{N_h} \omega(|\mathbf{R}_{\beta\gamma}|) (\delta_{\alpha\gamma} - \delta_{\alpha\beta}) \mathbf{I}^{(2)} \otimes \mathbf{R}_{\beta\gamma} \Delta V_\gamma \right) \cdot \mathbf{K}_\beta^{-1} = -\mathbf{I}^{(2)} \otimes \mathbf{D}_\beta, \quad \gamma \neq \beta. \quad (79)$$

To formula a single scale coarse grained MD, we first turn off the fine scale variables,

$$\chi_\alpha \approx \mathbf{I}^{(2)}, \quad \alpha = 1, 2, \dots, M$$

and for each cell,

$$\mathbf{S}_i \approx \mathbf{S}_i(0) = \mathbf{R}_{\alpha i}, \quad i = 1, 2, \dots, N_\alpha$$

We then obtain a coarse scale molecular dynamics,

$$M_\alpha \ddot{\mathbf{r}}_\alpha + \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} V'(r_{ij}) \frac{\mathbf{r}_{ij}}{|\mathbf{r}_{ij}|} + \mathcal{B}_\alpha = 0, \quad (80)$$

where

$$\mathbf{r}_{ij} = \mathbf{r}_{\alpha\beta} + \mathbf{F}_\beta \cdot \mathbf{R}_{\beta j} - \mathbf{F}_\alpha \cdot \mathbf{R}_{\alpha i}. \quad (81)$$

The coarse grain dynamic equations (80) and (81) form a close system.

To formula a two-scale coarse grained MD, we only turn off the fine scale oscillation,

$$\mathbf{S}_i \approx \mathbf{S}_i(0) = \mathbf{R}_{\alpha i}, \quad i = 1, 2, \dots, N_\alpha.$$

The governing equations of the two-scale molecular dynamics are,

$$M_\alpha \ddot{\mathbf{r}}_\alpha + \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} V'(r_{ij}) \frac{\mathbf{r}_{ij}}{|\mathbf{r}_{ij}|} + \mathcal{B}_\alpha = 0, \quad (82)$$

where

$$\mathbf{r}_{ij} = \mathbf{r}_{\alpha\beta} + \phi_\beta \cdot \mathbf{R}_{\beta j} - \phi_\alpha \cdot \mathbf{R}_{\alpha i}, \quad \text{where } \phi_\alpha = \chi_\alpha \cdot \mathbf{F}_\alpha. \quad (83)$$

The dynamic equation for micro-deformation tensor in each cell is determined by

$$\ddot{\phi}_\alpha \mathbf{J}_\alpha + \phi_\alpha (\mathcal{S}_\alpha^{int} - \mathcal{S}_\alpha^{ext}) \Omega_{\alpha 0} + \mathbf{M}_\alpha = 0, \quad (84)$$

with

$$\mathcal{S}_\alpha^{int} = \frac{1}{\Omega_{\alpha 0}} \sum_{i \in \alpha} \sum_{j \in \alpha, j \neq i} \frac{V'(r_{ij})}{r_{ij}} \mathbf{S}_i \otimes \mathbf{S}_i \quad (85)$$

$$\mathcal{S}_\alpha^{ext} = \frac{1}{\Omega_{\alpha 0}} \sum_{\beta \neq \alpha} \sum_{i \in \alpha, j \in \beta} \frac{V'(r_{ij})}{r_{ij}} \mathbf{r}_{ij} \otimes \mathbf{S}_i \quad (86)$$

and  $\mathbf{M}_\alpha = \sum_{i \in \alpha} \mathbf{f}_i \otimes \mathbf{R}_{\alpha i}$ . Eqs. (82)-(86) are also a closed system.

## VII. DISCUSSIONS

In this work, we have proposed in the first time a novel concept of multiplicative multiscale decomposition. By analyzing the structure of the (Andersen)-Parrinello-Rahman molecular dynamics, we have extended the (Andersen)-Parrinello-Rahman MD to form a novel multiscale micromorphic molecular dynamics (MMMD) that can solve finite size molecular dynamics problems without the restriction of the periodic boundary condition. In other words, it can solve finite size molecular dynamics problems with arbitrary boundary condition. This is because we can apply boundary conditions to the coarse scale variables, say  $\mathbf{r}_\alpha$ , to impose the boundary conditions at macroscale.

Different from the most multiscale methods proposed in recent years, the proposed multiscale dynamics formulation is not aimed at saving computation time or resource, but aimed at revealing multiscale connections and structures so that we can apply molecular dynamics to solve engineering problems with arbitrarily domain and general boundary condition. It is the author's opinion that if only we can achieve these goals we can start to think about how to build a coarse-grain model that can provide the efficient computing and save computational resources.

The conventional wisdom is that if we simply increase the size of molecular dynamics simulation we can simulate large and large size of objects based on the first principle. In order to capture correct thermodynamics response of a finite size molecular system, we cannot only solve massive numbers Newton equations, instead the system's multiscale characters must be carefully taken into account so that the microscale quantities can be correctly related to mesoscale and macroscale quantities based on first principle. It may be noted that the multiscale technique employed here is not for saving computer resource but for correct simulations of thermodynamic variables for a finite size system. Moreover, the MMMD formulation is essentially a local NSE ensemble formulation, and we have not considered the thermal or temperature effects yet. A future study to extend the present theoretical formulation to other molecular dynamics ensembles such as local NPT ensemble will be reported in a separated paper, and the computer implementation of the multiscale micromorphic molecular dynamics formulation will be reported in the second part of this work.

## Acknowledgments

The author would like to thank Professor Antonio DiCarlo for the enlightening discussions on the subject. Q. Tong is supported by a graduate fellowship from Chinese Scholar Council (CSC), and this support is greatly appreciated.

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